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I. Atomic physics

A. Ultra-cold collisions

The understanding of ultra-cold collisions played a crucial role in the achievement of Bose Condensation in a dilute vapor and in the understanding of the most important systematic effect in the creation of more precise atomic clocks. We have continued our work in these areas during this granting period.

1. Measurement of Collision Shift on Clock Transition and Potential Quantum Computation in Optical Lattices, C. Chin, V. Vuletic, A. J. Kerman and S. Chu, in *Laser Spectroscopy XV*, eds. S. Chu, V. Vuletić, A.J. Kerman and C. Chin (World Scientific, Singapore, 2002) pp. 317-320.

In this work, we explore how the collision shift on the atomic clock transition is a means of creating entangled states as a means of creating entangled states needed for quantum computation.

2. Sensitive detection of cold molecules by radiative Feshbach spectroscopy, C. Chin, A.J. Kerman, V. Vuletić, and S. Chu, *Phys. Phys. Rev. Lett.* **90**, 033201-3 (2003).

In this work, we observed the dynamic formation of quasi-bound Cs_2 molecules near Feshbach resonances in a cold cloud of atomic cesium. Using a new, ultra-sensitive photodissociation method, we have identified more than 15 weakly bound states with formation cross sections as small as 2×10^{-16} cm². We showed that more than 5×10^5 molecules can coexist with $\sim 10^8$ atoms.

3. Ultracold Cs₂ Feshbach Spectroscopy, C. Chin, V. Vuletic, A.J. Kerman, S. Chu, E. Tiesinga, P.J. Leo, and C.J. Williams, *Phys. Rev. A*, 70, 032701 (2004).

In this work, we report a summary of the more than 60 magnetic field-induced Feshbach resonances in ultracold collisions of ground-state cesium atoms. The detection is based on elastic, inelastic and radiative collision processes. The observations have allowed us to improve upon the interaction potentials between cesium atoms, which, in turn, allowed us to predict the possible magnetic field regions where the Bose condensation of cesium atoms would be possible. This knowledge provided the foundation for other researchers, who have achieved Bose condensation of cesium with the application of an external magnetic field in accordance with our predictions.

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B. Laser Cooling

4. Raman-sideband cooling in optical lattices, Andrew J. Kerman, Cheng Chin, Nathan Gemelke, Seokchan Hong, Keng Yeow Chung, Phillip Treutlein, Vladan Vuletić and Steven Chu, *Phys. Rev. A*, submitted (2004).

Raman sideband cooling methods capable of cooling atoms to extremely high phase space densities was demonstrated on cesium atoms, and discussed in our previous publications. This paper describes the full details of 3-D Raman sideband cooling and presents results of this cooling technique when applied to the cooling of rubidium atoms. The extension of our work to degenerate sideband cooling to Rb shows great promise for high-brightness applications where the collisional properties of Cs (very high collision cross-section) present limitations. In particular, rapid and efficient production of Bose-Einstein condensates may be aided by the use of a sideband pre-cooling stage, allowing subsequent evaporative cooling to initiate at much higher collision rates than possible with traditional recoil-limited cooling methods.

We have demonstrated both the efficient loading of molasses-cooled atoms into optical lattices, and the ability to cool these atoms near the recoil limit, at densities limited only by initial preparation of the sample. This has led to the production of phase-space densities three orders-of-magnitude larger than previously accessible through optical cooling. Once loaded and compressed in a typical performance magnetic trap, these sideband-cooled samples show promise for collision rates prior to evaporation an order-of-magnitude larger than possible with recoil-limited cooling methods. Typical performance figures for sideband-cooling in ⁸⁷Rb and ¹³³Cs are compared in the table for both two and three dimensional cooling, optimized for temperature and phase-space density.

Summary of $^{133}\mathrm{Cs}$ and $^{87}\mathrm{Rb}$ cooling results in near-detuned two and three-dimensional optical lattices.

	density n [atoms/cm ³]	temperature T [nK] (T/T_R)	$\begin{array}{c} \text{atom} \\ \text{number} \ N \end{array}$	Phase-space density
lowest ¹³³ Cs 3D temperature	3×10^{10}	290 (1.46)	1×10^8	
lowest ¹³³ Cs 2D temperature		210 (1.06)	1×10^8	
highest 133 Cs phase-space density	1.1×10^{11}	380 (1.92)	3×10^8	1/500
lowest ⁸⁷ Rb 3D temperature	6×10^{10}	500 (1.44)	2×10^8	
lowest ⁸⁷ Rb 2D temperature		250 (0.71)	1×10^8	
highest ⁸⁷ Rb phase-space density	3×10^{11}	750 (2.15)	5×10^8	1/350

C. Atom interferometry

We are finishing the final calculations that will be part of an atom interferometer measurement of h/M_{Cs} , which can then be used to obtain a value of the fine structure constant, α . Alpha can be written as

$$\alpha^2 = (\frac{2R_{\infty}}{c})(\frac{m_{p}}{m_{e}})(\frac{M_{Cs}}{m_{p}})(\frac{h}{M_{Cs}}),$$

where the Rydberg constant R_{∞} , and the mass ratios M_{Cs}/m_p , and m_p/m_e , have been determined with accuracies of 0.008, 0.20, and 2.1 ppb respectively. Thus, a measurement of h/M_{Cs} of comparable precision, in conjunction with the other measured quantities, yields an improved value of α .

The present accuracy is estimated to be \sim 4 ppb uncertainty, and progress reports of our work have been reported in several conference proceedings, refs. 5, 6a, and 6b listed below.

- 5. A measurement of the fine structure constant, Joel Hensley, Andreas Wicht, Edina Sarajlic and Steven Chu, eds. Steven Chu, Vladan Vuletić, Cheng Chin, Andrew Kerman in Laser Spectroscopy XV, eds. S. Chu, V. Vuletić, A.J. Kerman and C. Chin (World Scientific, Singapore, 2002) pp 133-142.
- 6a. A preliminary measurement of h/M_{Cs} with atom interferometry, Andreas Wicht, Joel Hensley, Edina Sarajlic and Steven Chu, Proceedings of the 6th Symposium on Frequency Standards, ed. P. Gill (World Scientific, Singapore, 2002).
- 6b. A preliminary measurement of the fine structure constant based on atom interferometry, Andreas Wicht, Joel Hensley, Edina Sarajlic and Steven Chu, Physica Scripta **102**, 82-88, (2002).
- 7. Localization effects in precision atom interferometry, A. Wicht, E. Sarajlic, J. M. Hensley, and S. Chu, Phys. Rev. A., submitted (2004).

We have delayed reporting a final number for h/M_{Cs} so that we can make theoretical estimates of two potential systematic effects. In this paper, we calculate the effects of having a localized cloud of atoms in a laser field of finite spatial extent. Previous to this work, treatments of precision atom interferometry were based on plane wave excitation of the atoms.

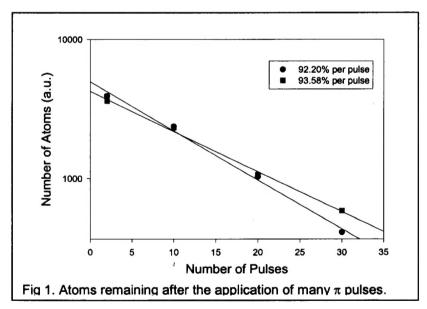
In another manuscript in preparation, we have calculated the systematic effect of momentum transfer of the optical pulse to the atoms in a cold atomic fountain due the collective presence of the cloud of cold atoms. This cloud affects the measured momentum transfer in two ways: (i) the index of refraction is no longer unity so the phase difference between the "up" and "down" interferometers that is used to deduce h/M_{Cs} is changed. (ii) The momentum imparted to the atoms by the optical field is changed.

To further reduce the uncertainty of the current measurement, we need to improve the signal to noise of our interferometer measurement. The measurement of h/M is based on the relative phase shift between two spatially separated atom interferometers that have been deflected in opposite directions by a number of π -pulses based on off-resonant Raman pulses. We have made good progress in developing a method to measure the phase shifts of both interferometers in the same atomic fountain launch. Noise common to both interferometers such as vibrations and microwave phase noise would be greatly reduced. In order to operate two atom interferometers simultaneously, we are constrained to use off-resonant Raman pulses. We have been able to improve the transfer efficiency of these pulses as shown in Fig. 1. By comparison, the previous best efficiencies of both the Chu and Kasevich groups were on the order of 75-80%.

We have run the two interferometers in the same launch, with the time separating the light pulses that address each interferometer equal to 1 ms. With this quasi-simultaneous operation, the noise was reduced by ~ 60 - 80%. The remaining noise is suspected to be due to detection noise (frequency and amplitude jitter of the laser used to read out the atomic populations after the completion of the interferometer pulses) and residual vibration noise due to the non-simultaneous operation of the two interferometers. For these reasons, we expect that the noise will be reduced further with true simultaneous interferometers and with improved detection.

Optical lattice sideband cooling methods that we developed during the last granting period have enabled us to launch a cloud of cold cesium atoms with temperatures ≤ 250 nK. Previously, we have made transitions between different hyperfine levels in order to insure that the atomic states could be frequency resolved. With modest additional velocity selection along

one dimension, we would have a sufficiently low velocity spread (without the added complication of using a Bose condensate) so that atom interferometers using the same internal atomic state but different external momentum states $|F, m_F=0; p\rangle$ and |F, $m_F=0$; p + 2n $\hbar k_{eff}$), where 2n is the number of photons used in the momentum exchange. We plan to increase the sensitivity of the interferometers by using beam splitters based on the exchange of 8 - 10 photon momenta



between atoms in the same internal state. Different atomic paths in the same internal state would decrease systematic phase shifts such as due to ac Stark shifts. Multiple photon transition beam splitters, if successful, would also be useful in creating "wide area" optical pulse interferometers for inertial sensing.

Assuming an initial atomic cloud temperature of ~ 150 nK, and a laser power of 200 mW in each beam, we could operate at a de-tuning of 10 GHz to the blue of the F=3 \rightarrow F'=4 transition. We calculate a two-photon Rabi frequency ~ 66 kHz and an on-resonant transition efficiency $\sim 90\%$ for a 10 photon transition. With these improvements, we can estimate the expected gain in precision. In our previous experiment, the fractional uncertainty in $\alpha = \Delta\alpha/2\alpha$ is given by the fractional phase noise $\Delta\phi/2\phi$

$$\frac{\Delta \alpha}{\alpha} = \frac{\Delta \phi}{2\phi} \approx \frac{40mrad}{32(N+1)n^2\omega_r T} \approx \frac{96.5}{(N+1)n^2T} ppb,$$

where N is the number of π -pulses, n is order of the transition, ω_r is the recoil frequency shift, and T is the time between $\pi/2$ pulses. For N = 30, T = 0.12 sec, n = 1, and 40 mrad phase noise per 40 launches, we had a 26ppb precision in one minute of integration time. Scaling from these results, for N = 4, n = 5, T= 0.2 sec, the precision would be 3.9 ppb in the same integration time. With an expected common mode noise improvement of 10, a relative phase noise of $\Delta \phi/2\phi \sim 0.39$ ppb in 1 minute integration time is expected. Even with \sim 4 ppb precision in one minute of integration time, we should be able to track down systematic effects at the 0.5 ppb level.

D. Other AMO work

8. Cold atoms and quantum control, Steven Chu, Nature 416, pp. 206-210 (2002).

A Nature Insight review article was the summary article to a collection of invited articles in a special issue of *Nature* discussing the applications of cold atoms. The article emphasizes that we now have unprecedented ability to control the quantum degrees of freedom of atoms and light. This control is linked to our ability to place atoms and photons into a well defined set of quantum states and to coherently manipulate these states. The *coherent* control of variables include the internal and external degrees of freedom of atoms, collisions between atoms, atomic de Broglie waves, spontaneous emission, many-body wave functions, and optical frequencies.

The papers listed below are mostly an indication of the PI's advanced age. The contributions include papers (and talks) that commemorated the retirement of his thesis advisor, Eugene Commins, the 60th birthday of a long time friend and colleague, Theodore Hänsch, and the passing of a long time friend and colleague, Arthur Schawlow.

- 9. A random walk in science, Steven Chu, in *Art and Symmetry in Experimental Physics, Festschrift for Eugene Commins*, eds. Dmitry Budker, Phillip, Phillip Bucksbaum, Stuart Freedman, (Am. Inst. of Physics, 2002) pp. 21-38.
- 10. High noise, low-resolution spectroscopy, in *Laser physics at the limits*, eds. H. Figger, D. Meschede, C. Zimmermann, (Springer, 2002) pp. 81-96.
- 11. Obituary for Arthur Schawlow for the National Academy of Sciences, Steven Chu and Charles Townes, (2003).

II. Polymer Physics

12. Visualization of molecular fluctuations near the critical point of the coil-stretch transition in polymer elongation", Hazen Babcock, Rodrigo Teixeira, Joe Hur, Eric Shaqfeh, and Steven Chu, Macromolecules 36(12), 4544 - 4548 (2003).

The behavior of flexible polymer molecules in two-dimensional fluid flows has been an area of active research for more than 30 years. All such flows could be divided into a mixture of elongational (stretching) (||E||) and rotational ($||\Omega||$) components. If ||E|| is greater than $||\Omega||$ neighboring fluid elements will separate exponentially with time. If ||E|| is less than $||\Omega||$ than they will never separate by more than some finite distance. The fluid elements exert hydrodynamic forces on the polymer as they separate. In the case where $||E|| > ||\Omega||$, as the fluid elements separate rapidly, the hydrodynamic force exerted on the polymer maybe large enough to overcome the entropic restoring force and cause the polymer to stretch. In contrast when $||E|| < ||\Omega||$, as the fluid elements never separate by more than a finite distance, they may not exert a strong enough hydrodynamic force on the polymer to overcome the entropic restoring force.

In 1974, de Gennes predicted that there would be a sharp coil-stretch transition in polymer behavior depending on the flow type and strength. In our earlier work, [Single Polymer Dynamics in an Elongational Flow, T.T. Perkins, D.E. Smith and S. Chu, Science **276**, 2016-2021 (1997); Response of Flexible Polymers to a Sudden Elongational Flow, D.E. Smith and S. Chu, Science **281**, 1335 (1998)], we were able to show that the sharp phase transition predicted by de Gennes was correct.

Previous experiments were plagued by variations in length in the polymer sample, and more significantly, by averaging over the behavior of many polymer molecules. By observing the behavior of single polymers in elongational flow, we discovered that identical molecules subject to the same external conditions would take different pathways to the extended equilibrium state, and that most of the molecules did not reach their extended equilibrium state during the time they were exposed to the elongational flow. Though our single molecule approach, the observed transition from coiled to extended state was roughly three orders of magnitude sharper than previously seen.

We have also studied shear flow which is a 50-50 mixture of elongation and rotation. [Single Polymer dynamics in Steady Shear Flow, D.E. Smith, H.P. Babcock and S. Chu, Science 283, 1724 (1999)] These studies showed that the polymers never reached an equilibrium state, but rotated and tumbled continuously and erratically.

De Gennes suggested that in flows close to shear, but with a slightly more elongational component, the coil-stretch phase transition may "soften" and for polymers subject to flows where $||E|| < ||\Omega||$, they should remain coiled.

Very long (E. Coli) DNA molecules were visualized in dilute solution using fluorescence microscopy. In planar extensional flow, polymer molecules were observed to exist in both coiled and highly-extended conformations over a narrow range of flow strengths. Polymer configuration hysteresis was observed for the first time following a prediction by de Gennes nearly 30 decades before. Polymer configurational free energy landscapes were calculated from computer simulations and show two free energy minima for flow strengths near the coil-stretch transition. Configuration hysteresis may directly influence bulk solution stresses and may have relevance to turbulent drag reduction and development of theoretical stress-strain relations for dilute polymer systems.

15. Effect of Hydrodynamic Interactions on DNA Dynamics in Extensional Flow: Simulation and Single Molecule Experiment, Schroeder, C. M.; Shaqfeh, E. S. G.; Chu, S.; Macromolecules 37, 9242-9256 (2004).

This paper quantified the role of hydrodynamic interactions in configuration hysteresis by comparing experimental observations to simulations.

16. The Dynamics of DNA in the Flow-Gradient Plane of Steady Shear Flow: Observations and Simulations, Charles M. Schroeder, Rodrigo E. Teixeira, Eric S. G. Shaqfeh, and Steven Chu, Macromolecules, in press (2004).

In this work, we introduced a novel method of observing the dynamics of single molecules of DNA in a shear flow in which the field of view is normal to the shear flow gradient plane. By exploiting the linear proportionality between polymer density and its recorded image, the accessible radius of gyration tensor elements were measured. For the first time, end-over-end tumbling was observed, confirming a long-standing prediction and numerous single-chain computer simulation studies. The tumbling frequency followed $Wi^{0.62}$, and an equation was derived from simple advection and diffusion arguments to reproduce these observations.

17. Biology and polymer physics at the single molecule level, Steven Chu, Phil. Trans. R. Soc. Lond. A **361**, 689-698 (2003).

This paper summarizes our most recent findings in single molecule polymer dynamics.

In this paper, we were able to observe the dynamics of polymers with 50.8% and 50.2% ||E|| component. We observed that the stretched polymers always align along the extensional eigenvector of the flow. The other eigenvector of the velocity matrix defines the direction of the compression axes for real eigenvalues of the velocity matrix. This result suggested that if we rescaled the results with the eigenvalue which gives the strength of the elongational flow, the data can be compared to that taken earlier in pure elongational flow experiments. The results of this work are

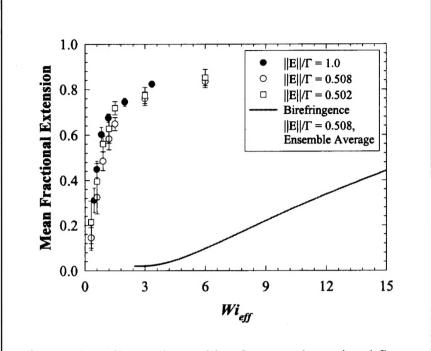


Fig. 1. The coil-stretch transition for pure elongational flows and flows close to pure shear conditions.

shown in Fig. 1 shows that the coil stretch extension of polymers in these flows remains very sharp.

The mean fraction extension plotted that is plotted in the Figure is the length of the polymer along the extensional eigenvector. For Weissenberg numbers very close to the critical gradient, Brownian fluctuations would flip the polymer onto the compressional eigenvector, causing the polymer to collapse and then re-extend along the extensional eigenvector.

We also examined the behavior of polymers when $||E|| = 0.495 ||\Omega||$. The polymers were seen to extend somewhat, but less than for the case of $||E|| = ||\Omega||$. Also, they showed a periodicity in the power spectral density spectrum equal to the rotation time of the flow.

13. The Dynamics and Configurational Fluctuations of Single DNA Molecules in Linear Mixed Flows, Joe S. Hur, Hazen Babcock, Steven Chu and Eric S.G. Shaqfeh, Phys. Rev. E 66, 011915 (2002).

Near the critical transition point between the coiled and stretched states of the polymer, large fluctuations are expected. This theoretical paper compares dynamical simulations with our experimental results.

14. Observation of polymer configuration hysteresis in extensional flow, Charles M. Schroeder, Hazen P. Babcock, Eric S.G. Shaqfeh, Steve Chu, Science 301, 1515-1519 (2003).

III. Biophysics

While the AFOSR grant was restricted to atomic physics and polymer dynamics, the work done in those areas had impact on the biophysics program. Below are listed the papers published or submitted in the previous granting period.

- 18. Initiation and re-initiation of DNA unwinding by the Escherichia coli Rep helicase, Taekjip Ha, Ivan Rasnik, Wei Cheng, Hazen P. Babcock, George Gauss, Timothy M. Lohman and Steven Chu, Nature 419, 638-641 (2002).
- 19. Channels in the Folding Landscape of a Structured RNA, Rick Russell, Xiaowei Zhuang, Hazen P. Babcock, Ian S. Millett, Sebastian Doniach, Steven Chu, and Daniel Herschlag, PNAS 99, 155-160 (2002).
- 20. Correlating Structural Dynamics and Function in Single Ribozyme Molecules, Xiaowei Zhuang, Harold Kim, Miguel J.B. Pereira, Hazen P. Babcock, Nils Walter, Steven Chu, Science **296**, 1473-1476 (2002).
- 21. Mg²⁺-Dependent Conformational Change of RNA Studied by Fluorescence Correlation on Immobilized Single Molecules, Harold D. Kim, G. Ulrich Nienhaus, Taekjip Ha, Jeffrey W. Orr, James R. Williamson and Steven Chu, PNAS 99, 4284-4289 (2002).
- 22. Exploration of the transition state for tertiary structure formation between and RNA helix and a large structured RNA, Laura E. Bartley, Xiaowei Zhuang, Rhiju Das, Steven Chu and Daniel Herschlag, J. Molec. Biology 328, 1011-1026 (2003).
- 23. Early steps of supported bilayer formation probed by single vesicle fusion analysis, Joseph M. Johnson, Taekjip Ha, Steve Chu and Steven G. Boxer, Biophysical Journal, 83, 3371-3379 (2002).
- 24. Single-molecule studies of SNARE complex assembly reveal parallel and antiparallel configurations, Keith Weninger, Mark E. Bowen, Steven Chu, and Axel T. Brunger, PNAS 100, 14800–14805 (2003).
- 25. Blanchard SC, Kim HD, Gonzalez RL, Jr., Puglisi JD, Chu S. tRNA dynamics on the ribosome. PNAS 101, 12893–12898 (2004).
- 26. Blanchard SC, Gonzalez RL, Jr., Kim HD, Chu S, Puglisi JD. tRNA selection and kinetic proofreading in translation, Nature Structural Biology 11, 1008 1014 (2004).
- 27. Single Molecule Observation of Liposome Bilayer Fusion Thermally Induced by SNAREs, Mark E. Bowen, Keith Weninger, Axel T. Brunger, and Steven Chu, Biophys. J., in press, (2004).
- 28. Single Molecule Studies of Synaptotagmin and Complexin Binding to the SNARE

Complex, Mark E. Bowen, Keith Weninger, James Ernst, Steven Chu and Axel T. Brunger, Biophys. J., submitted (2004).

Other Biophysics

- 29. Pulling on hairpins (Perspective) J. M. Fernandez, S. Chu and A.F. Oberhauser, Science **292**, 653-654 (2001).
- 30. Watching molecular systems work, one molecule at a time, Steven Chu, Proceeding to the Symposium on the Frontiers of Science, in celebration of C.N. Yang's 80th birthday (2003).
- 31. Single molecule studies of biological processes, Steven Chu, in *Atomic Physics 18*, eds. H.R. Sasdeghpour, E.L. Heller, D.E. Pritchard (World Scientific, Singapore, 2003) pp. 149-158.
- 32. Miniaturization in Biology, Steven Chu, Acumen Journal of Life Sciences (2004).

IV. Other activities

Talks and public outreach

Many plenary talks, invited talks and colloquia were given during this granting period by the PI, his post-docs and students. This included several plenary talks at AFOSR meetings and functions including a celebration of the 50th Anniversary of the AFOSR. The PI has also given many public talks, talks at summer camps and science fairs,

The P.I. testified on April 9, 2003 before the House Appropriations sub-committee for continued support of the physical sciences. The testimony was coordinated through the Joint Steering Committee for Public Policy.

There were two patents filed by Stanford: one on cavity cooling of atoms and molecules and one on the trapping and extension of single molecules of DNA in extensional flow.

Student and postdoctoral training in the atomic physics and polymer physics programs

Students who were trained and graduated during this granting period include Andrew Kerman (a postdoc who introduced Prof. David DeMille at Yale to laser cooling methods, and now a postdoctoral fellow with Wolfgang Ketterle, MIT), Ching Chen (formerly a postdoc with Vladan Vuletic, Rudi Grimm, and beginning as an Assistant professor at the University of Chicago), Heun-Jin Lee (currently a postdoc with Stephen Quake, Cal Tech), Joel Hensley (now working for Physical Sciences, Inc., Andover, MA), Hazen Babcock (a postdoc at Harvard) and Charles Schroeder (post doc at Harvard). Postdocs trained include Keng Yeow Chung (now an Assistant Professor at National University of Singapore), Vladan Vuletic (now an Associate Professor at MIT), and Andreas Wicht, (now an Assistant Professor at the Institut für Experimentalphysik, Heinrich-Heine-Universitaet Düsseldorf).